Determination of the long-term equilibrium factor indoors by means of Makrofol track-etched detectors (*)

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Summary. — From the point of view of radioprotection, it is worthwhile to determine the long-term equilibrium factor indoors to better ascertain the dose which population is exposed to. The method used in our laboratory to obtain long-term equilibrium factors indoors is based on simultaneous measurements of ^{222}Rn and ^{214}Po in air by using a detector with two *Makrofol-ED* (polycarbonate) foils covered with aluminised mylar: one placed inside a diffusion chamber, and the other one in direct contact with air, allowing, respectively, the measurement of ^{222}Rn and ^{214}Po activity concentrations. The equilibrium factor is estimated from the ratio of these two concentrations. The detector has been calibrated at the radon chamber of the National Radiological Protection Board (NRPB), Chilton, UK. Sensitivity values of $\varepsilon_0 = (0.85 \pm 0.04)$ (tr·cm^{-2})/(kBq·m^{-3}·h) and $\varepsilon_4 = (3.0 \pm 0.2)$ (tr·cm^{-2})/(kBq·m^{-3}·h) have been obtained from the calibration for the measurement of ^{222}Rn and ^{214}Po, respectively. A radon activity concentration of 44 Bq·m^{-3} and a mean equilibrium factor of 0.42 have been obtained in a pilot application indoors.

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1. – Introduction

The major contribution to the dose received by population is due to inhalation of the α-emissions from short-lived radon daughters. The European Union 96/29/EURATOM directive establishes, for the first time, the need of controlling radon levels in workplaces. Annual effective doses can be obtained from the Equilibrium Equivalent Radon concentration (EER) in Bq·m^{-3} and appropriate conversion factors [1]. The

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facts that i) radon is easier to measure than EER, and ii) radon and its daughters are not in equilibrium, have lead to the use of the equilibrium factor, F, defined as the ratio between the EER and the actual radon concentration, C_0. Therefore, EER can be calculated from equilibrium factor and radon concentration annual mean values.

Active systems are very useful to determine activity concentrations of radon and its daughters when the goal is to obtain their values at a specific time or to analyse their temporal evolution. However, due to their cost, size, and operating mode, they are not adequate for measurement of the annual average of EER in a significant fraction of dwelling stock. A passive integrating system such as Solid State Nuclear Track Detector (SSNTD) is more appropriate for this purpose. There is a great amount of data on indoor radon levels as a consequence of many surveys carried out around the world. The annual effective dose is estimated assuming a mean equilibrium factor of 0.45 [1]. Existing equilibrium factor data correspond to punctual or short-term measurements performed with active systems in few sites. The first studies about long-term measurements of the equilibrium factor with SSNTD were based on the simultaneous determination of the radon concentration with a closed detector and of the total alpha activity concentration in air, C_T, with a bare detector [2]. Recently, it was found that a more accurate estimation of the equilibrium factor can be obtained from the simultaneous measurement of the activity concentrations of ^{222}Rn, C_0, and ^{214}Po, C_4 [3]. In this paper, we establish a procedure for determining the long-term equilibrium factor based on the last method, using *Makrofol-ED* detectors. Results from calibration and from a pilot application indoors are presented as well.

2. – Principles of the equilibrium factor estimation with SSNTD

The equilibrium factor can be calculated from the short-lived radon daughters activity concentrations as follows:

$$F = \frac{0.104\,C_1 + 0.516\,C_2 + 0.380\,C_3}{C_0}\,, \tag{1}$$

where C_1, C_2, and C_3 are the activity concentrations of the ^{218}Po, ^{214}Pb and ^{214}Bi, respectively.

Radon daughters behaviour in air is much more complicated than radon behaviour because of their interaction with aerosol particles. The basic processes that influence the activity balance of radon daughters and that break the equilibrium with radon are the air exchange, the surface deposition, and the attachment to aerosol particles. Assuming a steady state, each radon daughter concentration, C_j, can be related to its parent concentration, C_{j-1}, by the following expression:

$$C_j = \frac{\lambda_j C_{j-1}}{\lambda_j + \lambda_v + h_j \lambda_d^a + (1 - h_j)\cdot\lambda_d^f}\,, \tag{2}$$

where λ_j (h^{-1}) is the decay constant of the j-th nuclide, λ_v (h^{-1}) is the ventilation rate of the room, the quantities λ_d^a and λ_d^f (h^{-1}) are the attached and unattached deposition rates, respectively, and h_j is the attachment fraction of the j-th radon daughter.

As a first approximation, we consider only the ventilation rate as free parameter and we assume a value of 0.2 h^{-1} and 20 h^{-1} for the attached and unattached deposition

rates, respectively, and a value of 0.9 for the attachment fraction of the ^{218}Po. These are usual values taken from the literature [2, 4]. We have also supposed that the remaining radon daughters cannot be found in unattached form ($h_2 = h_3 = h_4 = 1$). Using all these values and defining the disequilibrium degree k_j of each radionuclide j as the ratio between its activity concentration and that of radon, we obtain from eq. (2)

$$k_1 = \frac{13.64}{15.82 + \lambda_v}, \tag{3a}$$

$$k_2 = \frac{21.14}{(1.75 + \lambda_v)\cdot(15.82 + \lambda_v)}, \tag{3b}$$

$$k_3 = \frac{44.61}{(2.31 + \lambda_v)\cdot(1.75 + \lambda_v)\cdot(15.82 + \lambda_v)}, \tag{3c}$$

$$k_4 = k_3. \tag{3d}$$

Equation (3*d*) is a consequence of the fact that ^{214}Bi and ^{214}Po are in secular equilibrium. From each value of the ventilation rate, λ_v, we obtain the corresponding values of the disequilibrium degree, k_j, and of the equilibrium factor, so that it is possible to determine the equilibrium factor from the values of k_j. In fig. 1, we have plotted the equilibrium factor as a function of k_j and of the ratio C_T/C_0 ($C_T = C_0 + C_1 + C_4$) as well. We can see in the figure that the equilibrium factor can be estimated if one of the disequilibrium degrees or the ratio C_T/C_0 is measured. Since β-particles cannot be registered with SSNTDs, k_2 cannot be determined and hereafter we will consider only the term k_4, which corresponds to ^{214}Po. The energy of α-particles emitted by ^{218}Po (6.00 MeV) is smaller than that of α-particles emitted by ^{214}Po (7.69 MeV). Any etching conditions allowing the registration of ^{218}Po α-particles will register at least a fraction of ^{214}Po α-particles as well, so that it is impossible to determine k_1 with track-etch detectors. Therefore, only the functions of $F = f(C_T/C_0)$ and $F = f(k_4)$ remain as functions of interest. It can be observed that the curve of $F = f(C_T/C_0)$ presents a more

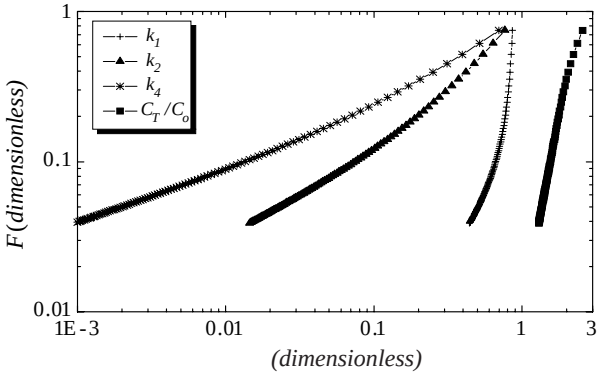

Fig. 1. – Equilibrium factor as a function of disequilibrium degrees for ^{218}Po (k_1), ^{214}Pb (k_2) and ^{214}Po (k_4), and of the C_T/C_0 ratio.

pronounced variation than the curve of $F = f(k_4)$. Assuming a relative uncertainty of 10% for the k_4 measurement, a relative uncertainty of about 5–10% in the estimation of the equilibrium factor value is obtained; while on the other hand, if we consider the same uncertainty for the C_T/C_0 value, the equilibrium factor uncertainty will be about 25–50%. As a consequence, if we optimise the experimental conditions for the k_4 determination, the equilibrium factor can be estimated from the $F = f(k_4)$ curve with the best possible precision.

3. – Experimental method

To determine experimentally the long-term disequilibrium degree k_4, the detector system must measure simultaneously the activity concentration of ^{222}Rn and of ^{214}Po. With this purpose, we have built a detector consisting of two *Makrofol-ED* foils covered with aluminised mylar: one 300 μm thick enclosed within a diffusion chamber, that allows the measurement of the radon concentration C_0, and a second one, 490 μm thick, bare, that permits the detection of the α-particles emitted by the ^{214}Po. Figure 2 shows the schematical representation of the detector. Standard etching conditions for the radon measurement, as found by our group [5], are used in the case of the 300 μm closed foil. In the case of the bare detector, a 41 μm layer must be removed during a pre-etching process in order to eliminate the registration of α-particles other than those emitted by the ^{214}Po [6]. In consequence, there is risk of perforation during electrochemical etching if 300 μm thick foils are used. A thickness of 490 μm has been found appropriate for the bare *Makrofol-ED* foil.

To avoid the ^{218}Po and ^{214}Po plate-out effects on the detector readings, the etching conditions have been optimised to detect an energy range of 6.1–7.5 MeV, and the

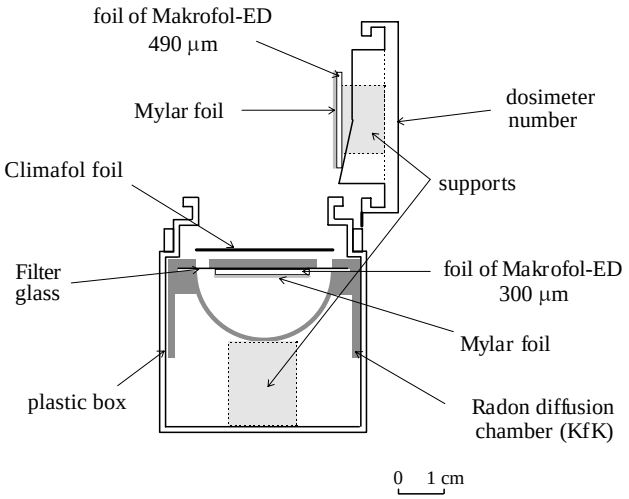

Fig. 2. – Schematic representation of the detector used for determinig ^{222}Rn and ^{214}Po activity concentration.

etching conditions for the ^{214}Po detection are [6]

pre-etching:

- Temperature: 40 °C
- Etching solution: KOH 7.5 N (1 : 1) ethanol-96%
- Duration: 6 h
- Bulk etch rate: (7.79 ± 0.08) $\mu m \cdot h^{-1}$

electrochemical etching:

- Temperature: 40 °C
- Duration: 1 h
- Frequency: 3 kHz
- Voltage: 1500 V
- Bulk etch rate: (8.9 ± 1.1) $\mu m \cdot h^{-1}$

Track counting of bare and closed *Makrofol-ED* foils is performed with the semi-automatic system available in our laboratory [7]. Twenty detectors were exposed at the National Radiological Protection Board (NRPB) Radon Environmental Chamber at three different concentrations of ^{222}Rn and of ^{214}Po for calibration purposes. The sensivity values, calculated by weighted least-squares fit to the experimental data, are

$$\varepsilon_0 = (0.85 \pm 0.04)\left(\frac{tr \cdot cm^{-2}}{kBq \cdot m^{-3} \cdot h}\right) \quad \text{for } ^{222}Rn, \tag{4}$$

$$\varepsilon_4 = (3.0 \pm 0.2)\left(\frac{tr \cdot cm^{-2}}{kBq \cdot m^{-3} \cdot h}\right) \quad \text{for } ^{214}Po. \tag{5}$$

4. – Results

As a pilot application indoors, 10 detectors were exposed for three months in a ground floor room in house located in the Barcelona area with normal ventilation and in which their occupants spent all the working day. A radon activity concentration of 44 $Bq \cdot m^{-3}$ and a mean equilibrium factor of 0.42 have been obtained. These values agree with previous radon surveys in the Catalan region [8] and with the world mean value of 0.45 for the equilibrium factor estimated by the International Commission on Radiological Protection (ICRP) [1].

5. – Conclusions

A detector consisting on two *Makrofol-ED* nuclear track foils designed to measure simultaneously the activity concentration of the ^{222}Rn and ^{214}Po in air has been built and calibrated. The equilibrium factor is estimated from the disequilibrium degree of ^{214}Po with the best possible precision allowed by the track-etch methodology. Preliminary measurements indoors show that the radon levels obtained agree with previous surveys in the region and that the equilibrium factor estimated is similar to the world averaged value.

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